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Wigner Supersolid of Excitons in Electron-hole Bilayers

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Bilayer electron-hole systems, where carriers in one layer are electrons and carriers in the other are holes, have been actively investigated in recent years with the focus on Bose-Einstein condensation of excitons. This condensation is expected to occur when the layer separation d is much smaller than the interparticle distance $r_s a_B$ within each layer. In this note, we argue for the existence of a state, Wigner supersolid, in which excitons are phase-coherent but form a Wigner crystal due to dipolar repulsion. We present the qualitative phase diagram of bilayer system, and discuss properties and possible signatures of the Wigner supersolid phase.

Introduction: The phenomenon of Bose-Einstein condensation (BEC) where the many-body wavefunction for the ground state of a macroscopic number of bosons is described by a uniform phase and acquires phase rigidity, is a remarkable manifestation of interplay between quantum mechanics and statistics of the particles. This phenomenon does not depend on the microscopic structure of bosons or their interactions, and does not make assumptions about possible broken symmetries in the ground state. Based on these observations, Moskalenko, Blatt, and Keldysh predicted that excitons - metastable bound states of electron-hole pair - in semiconductors will undergo BEC under appropriate circumstances.[1] Electron-hole bilayers are expected to exhibit a *uniform* BEC of excitons - dipolar superfluid - when the distance between the two layers d is much smaller than the typical interparticle distance $r_s a_B$.[2,3,4] Here $a_B = \hbar^2\epsilon/m^* e^2$ is the Bohr radius for a particle with band mass m^* in a semiconductor with dielectric constant ϵ (For typical semiconductors, $\epsilon \sim 13$ and $m^* \sim 0.10\, m_e$ implies that $a_B \sim$ 100 Å). In recent years, advances in sample preparations have made it possible to fabricate samples in which the two layers are close ($d \sim$ 100-300 Å), the exciton lifetime is relatively long, and the carrier mobilities at low densities are high or, equivalently, disorder effects are small.[5,6,7,8,9] Therefore, an experimental exploration of the entire phase-diagram of electron-hole bilayer system will be feasible in near future.[10]

Recent experiments have sparked interest in two disparate aspects of BEC, namely its realization in semiconductors and in solid Helium under pressure.[11] These two aspects, condensation of (metastable) bosons in a strongly interacting semiconductor environment and non-uniform Bose-Einstein condensates, address fundamental questions such as what are necessary and sufficient conditions for Bose-Einstein condensation? Is superfluidity related to a *uniform* Bose-Einstein condensate? *In this note, we show that electron-hole bilayers support a ground state which combines these two features, namely Bose-Einstein condensation and broken translational symmetry.*

In the following, we first discuss zero-temperature phase diagram of electron-hole bilayer system. Then we present qualitative description of various phase boundaries and focus on the transition from the dipolar superfluid phase to the Wigner supersolid phase. We end the note with discussion regarding experimental signatures of the Wigner supersolid phase and conclusions.

Phase Diagram: Let us consider a bilayer system with electrons in the top layer and holes in the bottom layer. We choose a convention such $e^\dagger(\mathbf{r})$ denotes an operator which creates electron at position $\mathbf{r}$ and $h^\dagger(\mathbf{r})$ denotes an operator which creates a hole at position $\mathbf{r}$ in the bottom layer. The density matrix of this system has four components. $\rho_{ee}(\mathbf{r},\mathbf{r}') = \langle e^\dagger(\mathbf{r})e(\mathbf{r}')\rangle$ and $\rho_{hh}(\mathbf{r},\mathbf{r}') = \langle h^\dagger(\mathbf{r})h(\mathbf{r}')\rangle$ denote density matrices for electrons and holes, and $\Delta(\mathbf{r},\mathbf{r}') = \langle e^\dagger(\mathbf{r})h^\dagger(\mathbf{r}')\rangle = \Delta(\mathbf{r}',\mathbf{r})^*$ denotes the interlayer phase-coherence matrix elements. This system is characterized by two dimensionless parameters which can be tuned independently. The first, $d/(r_s a_B)$, is the ratio of intralayer and interlayer Coulomb interactions (PE/PE2), and is a measure of "phase coherence" between the two layers. This parameter, for $d/(r_s a_B) < 1$, drives the quantum phase transition from uncorrelated bilayers, $\Delta = 0$, to a state with phase-coherent bilayers, $\Delta \neq 0$. We emphasize that here we have assumed the simplest scenario, in which the formation of individual excitons and the establishment of long-range phase coherence happen simultaneously. In a more general case, there will be two phase transitions, one corresponding to each of the two possibilities outlined above. The second parameter r_s is the ratio of potential energy $e^2/\epsilon(r_s a_B)$ and kinetic energy $\hbar^2/m^*(r_s a_B)^2$ of carriers within a single layer (PE/KE). This parameter, for $r_s \gg 1$, drives the quantum phase transition from a uniform density state, $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) \propto \delta_{\mathbf{q},0}$, to the Wigner crystal state with broken translational symmetry, $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) \propto \delta_{\mathbf{q},\mathbf{G}}$ where $\mathbf{G}$ is a reciprocal lattice vector for Wigner crystal. Therefore, in the simplest scenario, we expect $2^2 = 4$ possible distinct ground states (Figure 1). For small $d/(r_s a_B)$ and small r_s ($d/a_B \leq r_s \leq 1$), the ground state of the system is a uniform Bose-Einstein condensate of excitons or a dipolar superfluid.[12,13] At large $d/(r_s a_B)$ and small r_s, the ground state is a uniform 2-component

(electron-hole) plasma. At large values of $d/(r_s a_B)$ and large r_s ($d/a_B \gg r_s \gg 1$) the system consists of Wigner crystals in respective layers which are weakly correlated. These three phases have been considered in the literature.[12] In particular, excitonic superfluid and Wigner crystal, being broken symmetry states, have been extensively studied. For example, spontaneous interlayer coherence has been observed in quantum Hall bilayers near total filling factor one[8] although compelling evidence for true off-diagonal long-range superfluid order is still somewhat ambiguous. Similarly, it is widely believed[14] that a single electron (or hole) layer becomes a Wigner crystal for large $r_s = r_{s0} \sim 40$ although the experimental evidence is not conclusive.

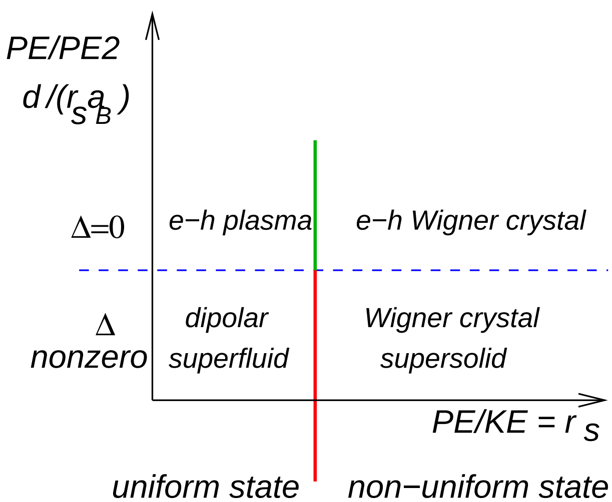

FIG. 1: Schematic phase diagram of a system with two dimensionless parameters $d/(r_s a_B)$ and r_s, showing four possible phases. We emphasize that the details of the topology, for example, the dependence of Wigner crystallization threshold r_s on the ratio of intralayer-to-interlayer potential energy $d/(r_s a_B)$, are not known. Therefore, the intersection of the two lines should not be considered a multi-critical point. The four possible ground states are i) uniform dipolar superfluid ii) electron-hole plasma iii) uncorrelated Wigner crystals, and iv) phase-coherent bilayers with broken translational symmetry. The first three have been extensively discussed in the literature.

In this note, we focus on the fourth possible ground state, which *will* occur when $d/(r_s a_B) \leq 1$ and $r_s \gg 1$. We point out that the two broken-symmetry states - dipolar excitonic condensate and Wigner crystal - will coexist in this regime. Based on general principles, this phase will have a ground state with phase-coherence between the two layers and a broken translational symmetry: $\Delta \neq 0$ and $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) \propto \delta_{\mathbf{q},\mathbf{Q}}$. *We propose that this state is a Wigner crystal of phase-coherent excitons: a Wigner supersolid.*

Now, we motivate the existence of this phase and show how various phase boundaries can be qualitatively understood. Figure 2 shows the (same) phase diagram of a bilayer electron-hole system[12] as a function of d/a_B and r_s (solid lines only). We choose these variables, instead of the ratio of energies, as the axes because experimentally these two can be tuned independently. This phase diagram does not take into account the transition from phase-coherent bilayers ($\Delta \neq 0$) to uncorrelated bilayers ($\Delta = 0$) which happens at $d/a_B \sim r_s$ (dotted blue line). Let us consider the region between the solid (green and red) lines from two different limits, increasing d/a_B at moderate r_s and increasing r_s at moderate d/a_B. First, we start from the uniform dipolar superfluid state characterized by uniform phase coherence $\Delta \neq 0$ and uniform density $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) = n_0 \delta_{\mathbf{q},0}$. The kinetic energy per exciton[15] is given by $\hbar^2/m^*(r_s a_B)^2$ whereas the potential energy due to dipolar repulsion is given by $e^2 d^2/\epsilon(r_s a_B)^3$. Therefore, when $\sqrt{r_s} \leq d/a_B$ potential energy dominates the kinetic energy and the excitons will form a hexagonal Wigner crystal[16,17] to minimize the potential energy. This argument gives the phase boundary between the uniform dipolar superfluid state, and the Wigner crystal of excitons characterized by $\Delta \neq 0$ and $\rho_e e(\mathbf{q}) = \rho_h h(\mathbf{q}) \propto \delta_{\mathbf{q},\mathbf{G}}$ (solid red line). *Such a phase with dipolar exciton condensate and crystalline structure within each layer is, by definition, a supersolid.* Since this is a transition from a uniform state to a crystalline state, in the absence of disorder, it will be a *first-order*

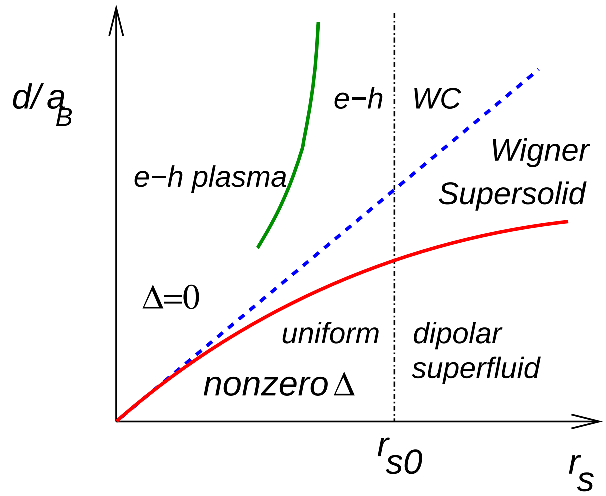

FIG. 2: Schematic phase diagram of the electron-hole bilayer. Standard phases discussed in the literature are electron-hole Wigner crystal ($d/a_B \gg 1$, $r_s \gg 1$), 2-component plasma ($d/a_B \gg 1$, $r_s \leq 1$) and dipolar superfluid ($d/a_B \leq 1$, $r_s \gg 1$). We postulate that a Wigner crystal of phase-coherent excitons exists in the region $\sqrt{r_s} \leq d/a_B \leq r_s$, between the dipolar superfluid and the uncorrelated Wigner crystals. We call this phase Wigner supersolid (WS) because it shows broken translational symmetry in the diagonal density matrix elements, $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) \propto \delta_{\mathbf{qG}}$, as well as interlayer phase coherence, $\Delta \neq 0$, in the off-diagonal density matrix elements. The transition between dipolar superfluid and WS is of first order as any liquid-to-solid transition; on the other hand, the transition between WS and electron-hole Wigner crystal is of second order as it is associated with the disappearance of phase coherence.

transition.

The existence of this phase can also be argued if we start with system in the uniform 2-component plasma state characterized by $\Delta = 0$ and $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) = n_0\delta_{\mathbf{q},0}$. As r_s increases, each layer undergoes hexagonal Wigner crystallization at a critical value of $r_s(d/a_B)$, leading to a ground state with no phase coherence, $\Delta = 0$, but crystalline density modulations, $\rho_{ee}(\mathbf{q}) = \rho_h h(\mathbf{q}) \propto \delta_{\mathbf{q},\mathbf{G}}$.[14] At large $d/a_B \gg 1$, this value of r_s is roughly independent of the value of d/a_B, and will approach the single-layer value $r_{s0} \sim 38$ asymptotically (solid green line). The behavior of this phase boundary at small $d/a_B > r_s$ will, in general, be nontrivial, due to interlayer interactions playing a role in determining the ground state. Now as d/a_B is reduced or, equivalently, r_s is increased, when $d/a_B \leq r_s$ (dotted blue line), the two Wigner crystals will become phase-coherent, $\Delta \neq 0$, and will still maintain the broken translational symmetry, $\rho_{ee}(\mathbf{q}) = \rho_{hh}(\mathbf{q}) \propto \delta_{\mathbf{q},\mathbf{G}}$, thus forming a hexagonal Wigner crystal of phase-coherent excitons.[14,16,17] This transition between two layers with identical crystal structure become is associated with the appearance of phase coherence and is therefore *continuous*.

We emphasize that this novel phase, a Wigner supersolid, is possible only due to specific properties of electron-hole bilayers. Traditionally, supersolid phase discussed in the literature has been mostly in the context of ^{4}He.[11] Some recent work has introduced the possibility of a supersolid phase in cold-atom optical lattice systems with extended Hubbard interactions[18] as well as in lattice models of hard-core bosons with repulsion.[19] Since excitons in bilayer systems have dipolar repulsion, which provides the incentive for localization, a Bose-Einstein condensate with broken translational symmetry is possible. In a single layer system, carriers undergo Wigner crystallization but since they are fermions there is no phase coherence. On the other hand, bulk semiconductors support excitonic condensation. However, since the exciton interaction is not necessarily repulsive (due to random orientation of exciton dipoles), there is no Wigner crystallization. Electron-hole systems, in which all excitons have the same dipole moment, offer a unique realization of hard-core bosons with repulsive interactions in a semiconductor environment.

Now we sketch a simple model for phase transition from the uniform dipolar superfluid phase to the Wigner supersolid phase. The dipolar superfluid is characterized by a nonzero order parameter $\Delta = |\Delta|\exp[i\Phi]$ where Φ is the dipolar phase. Note that the density of this superfluid is uniform. The long-wavelength low-energy dynamics of a dipolar superfluid is described by the action[13]

$$\mathcal{S}_0 = \sum_{\mathbf{k},\omega}\left[\omega\rho_{\mathbf{k}\omega}\Phi_{\mathbf{k}\omega} - \frac{\rho_d}{2}k^2\Phi^*_{\mathbf{k}\omega}\Phi_{\mathbf{k}\omega} + \frac{1}{2}S^{-1}_{\mathbf{k}\omega}\rho^*_{\mathbf{k}\omega}\rho_{\mathbf{k}\omega}\right]. \tag{1}$$

Here, ρ_d is the superfluid phase stiffness, $\rho_{\mathbf{k}\omega}$ is the Fourier transform of the dipolar density fluctuation, and the dipolar phase and condensate number satisfy $[\rho, \Phi] = i$. We introduce $S^{-1}_{\mathbf{k}\omega}\rho^*_{\mathbf{k}\omega}\rho_{\mathbf{k}\omega}$ term in the action to account for the dipolar density-density interaction. At small wavevectors, this interaction gives the capacitive mass-term for uniform density fluctuations. Integrating the density fluctuations, we arrive at the action for the dipolar phase,

$$\mathcal{S}_\Phi = \frac{1}{2}\sum_{\mathbf{k},\omega}(S_{\mathbf{k}\omega}\omega^2 - \rho_d k^2)\Phi^*_{\mathbf{k}\omega}\Phi_{\mathbf{k}\omega}. \tag{2}$$

The dispersion of the collective mode is given by $\omega_{\mathbf{k}} = k\sqrt{\rho_d/S_{\mathbf{k}\omega}}$. When $k \to 0$, this gives the linearly dispersing sound mode, $\omega_{\mathbf{k}} = k\sqrt{\rho_d/C} = v_c k$, where $C = S_{\mathbf{k}=0,\omega=0}$ is the capacitance. Since the dipolar fluid is a uniform Bose-Einstein condensate, the phase collective mode will have a roton minimum at a wavevector inversely proportional to the interparticle distance.[17,20] In the vicinity of this minimum, for $\mathbf{k} \simeq \mathbf{k}_0$, the dispersion will be gapped with $\omega_{\mathbf{k}} \simeq \Delta_r + (\mathbf{k}-\mathbf{k}_0)^2/2m_r$ where Δ_r is the energy gap at the roton minimum and m_r is the roton mass. We postulate, based on results for collective mode dispersion in similar systems,[17,21], that as d/a_B increases the roton gap Δ_r is suppressed, reflecting the tendency towards a state with a broken translational symmetry with characteristic wavevector $k_0 = (r_s a_B)^{-1}$. It follows (from the existence of a ring of roton modes which are softening) that the system "jumps" into Wigner supersolid phase via a first-order phase transition, as it does in the case of liquid-to-solid transition in ^{4}He under pressure.[22] Similarly, we postulate that the roton gap remains finite when the crystalline order emerges, as it does in ^{4}He and in quantum Hall bilayers; the magnitude of the roton softening can only be addressed by a microscopic calculation.[17]

How to detect a Wigner Supersolid? Observation of a Wigner supersolid, with two nonzero order parameters, will require separate measurements of phase coherence and crystalline order. Let us consider how the observable properties change when we enter Wigner supersolid phase from the uniform dipolar superfluid phase or the electron-hole Wigner crystal phase. The uniform dipolar superfluid phase is characterized by dissipationless counterflow currents proportional to the in-plane magnetic field,[13] increased exciton recombination rate,[5,6,7] and, because the excitons are delocalized, an enhanced interlayer tunneling conductance,[8] When the system becomes supersolid, the phase coherence will be manifest in enhanced recombination rate and dissipationless counterflow. However, since the excitons are localized, interlayer tunneling conductance will be *suppressed* compared to its value in the superfluid phase. Since the exciton recombination in the Wigner crystal is restricted to lattice sites, *Fourier transform of spatially resolved photoluminescence will reflect the crystalline structure.* The uncorrelated Wigner crystal phase is characterized by insulating behavior for in-plane and interlayer transport, as well as phonons with dispersion $\omega_p \propto k^{1/2}$ since the Coulomb interaction is $V(r) = e^2/\epsilon r$. When the two Wigner crystals become

phase coherent and the system becomes a supersolid, the in-plane transport will support dissipationless counterflow, and the phonon dispersion will change to $\omega_p \propto k^{3/2}$ since the dipolar interaction is $V_d(r) = e^2d^2/\epsilon r^3$. In addition, the interlayer tunneling conductance will be *enhanced* compared to its value in the electron-hole Wigner crystal state, due to increased recombination rate.

Thus, the existence of a Wigner supersolid can be confirmed by transport measurements (interlayer tunneling conductance, counterflow superfluidity) and spatially resolved photoluminescence measurements.

Conclusion: We have shown, based on general principles, the existence of a supersolid phase in low-density electron-hole bilayers with moderate interlayer separations. Since the critical temperature for BEC in two dimensions is $T = 0$, we have focused on quantum phase transitions between possible ground states of this system in the absence of disorder; thus, our discussion is only applicable at low temperatures and to ideally pure systems. Experimental observation of the proposed supersolid phase will require studying the effects of disorder and finite temperature, which are beyond the scope of this note. We believe that at finite temperatures and in the presence of finite disorder, our proposed Wigner supersolid is likely to be a crossover phase with the strict supersolid existing only at $T = 0$. More theoretical work will be needed to address this question.

We have shown that experimental observation of the Wigner supersolid will require separate probes looking at the phase-coherence response and intralayer Wigner crystal response. Such a supersolid behavior should exhibit strong dependence on layer separation, temperature, and r_s since, as we have shown, the supersolid phase will only be stable in the region $\sqrt{r_s} \leq d/a_B \leq r_s$, at low temperatures. Although the experimental observation of our proposed Wigner supersolid may be difficult, we believe that its existence in electron-hole bilayers is a robust conclusion. The exact values of d/a_B and r_s at which Wigner supersolid is realized will have to be determined via Monte Carlo simulations.[17] Wigner crystallization in electron-hole bilayers occurs[12] at $r_s \sim 20$ compared with[14] $r_s \sim 40$ in the single-layer system and is thus energetically favorable in bilayer systems. This observation makes the case stronger for the existence of our proposed Wigner supersolid phase.

Wigner supersolid of excitons in semiconductors is a natural extension of the excitonic condensate, just as a supersolid is a natural extension of Bose-Einstein condensate with repulsive interactions.[16,17,18,19]. It will be interesting to explore the consequences of a similar analysis in electron-electron (or hole-hole) bilayers where spontaneous interlayer phase coherence may exist[23] in the absence of a magnetic field, as well as in the case of quantum Hall bilayers[24]. The verification (or falsification) of our proposed Wigner supersolid will further our understanding of the interplay between interparticle interactions and quantum statistics.

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